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SEMIANNUAL REPORT

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Measurements of the fluorescent spectral intensity and predictions of stimulated emission cross sections for the $NaXe_2$ molecule have been placed on an absolute basis. Numerical models are being developed to describe the formation of the cathode fall region of high power electrical discharges, such as used for visible lasers. Measurements of electron excitation rate coefficients for the $b^1\Sigma_g^+$ state of O_2 show that at the higher mean energies the production of $b^1\Sigma$ is dominated by collision induced cascading from higher excited state. Measurements of the decay of the Na resonance radiation have been (continued)

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interpreted in terms of the collision transfer of excitation to and from the $A^1\Sigma_u^+$ and $a^3\pi_u$ states of Na_2.

SEMIANNUAL REPORT

This Semiannual Report contains descriptions of work carried out under ONR Contract No. N00014-76-C-0123 and ARPA Order No. 2683, and Amd. 3, and covers the period from 1 February 1976 to 31 July 1976. Section I is the Semiannual Report Summary while Sections II-V are more detailed descriptions of work carried out under the four projects supported by this contract.

I. SEMIANNUAL REPORT SUMMARY

The four projects being carried out in the area of Laser Processes under this contract are summarized below. More detailed discussions are given in Sects. II through V of this report.

1) Generation and Interpretation of Molecular Continuum Radiation

Many metal-noble gas molecules are viable condidates for high-efficiency, high-power excimer lasers. Since the spectra and potentials of most of these molecules are unknown, the merits of different metal-noble gas combinations can only be assessed from detailed measurements of the kind reported here. The alkali-noble gas molecules, which we have studied in detail under this and previous contracts, are candidates for excimer lasers in the 0.7-1 μm wavelength region. Since the proposed laser systems operate best at many atmospheres of buffer gas, we are measuring the increases in the stimulated emission coefficients which occur when the alkali atom-rare gas excimers are perturbed by a second rare gas atom. Thus, in this Semiannual Technical Report we summarize measurements of the absolute values of the fluorescent efficiencies for the triatomic molecules $NaXe_2$ which at 20 atm total pressure dominate the stimulated emission in Na-Xe mixtures at wavelengths greater than 720 nm. Comparisons of the experimental fluorescence data with the predictions of a rather simple model of the $NaXe_2$ molecule show agreement to within a factor of two. This means that we have developed techniques for making good estimates of the role of triatomic molecules in the proposed high power laser system.

2) Stability of Discharges in Weakly Ionized Gases

The technical problem being investigated is the optimization of the use of electrical energy for the production of excited molecules in electrically excited gas lasers. In particular, the efficient use of electrical excitation in gas lasers requires that the discharge remain diffuse as the density of the excited molecules is raised as high as possible, i.e., that the discharge not form a constricted channel or arc. During this report period we have concentrated on the development of numerical models of the cathode fall region of high-power laser discharges. Since the cathode fall is the portion of a glow discharge with the highest energy input per unit volume it is considered by many to be the ultimate source of the runious arcs which generally limit the output of high-power, electric-discharge lasers.

3) Electron Excitation of Molecular Metastables

The electron excitation of molecular radiators in an electrical discharge can be an efficient means of producing high-power densities in large lasers. During this report period we have completed measurements of the rate coefficients for excitation of the $b^1\Sigma_g^+$ state of O_2 by electrons with mean energies between 0.75 and 6 eV, i.e., the range of mean energies of interest in electrical discharge lasers. Our results show that collisions of excited O_2 molecules and $O(^1D)$ atoms produced by dissociation excitation of O_2 with ground state O_2 are very efficient sources of $O_2(b^1\Sigma_g^+)$ molecules, e.g., the net efficiency of excitation of $b^1\Sigma$ is greater than 20% for mean electron energies between 1.5 and 6 eV.

4) Scattering and Transport of Resonance Radiation in Gases

The technical problem considered here is the measurement of the transport and scattering of resonance radiation emitted by metal vapors so as to obtain the rates of radiative and non-radiative energy loss by excited atoms in excimer lasers, etc. The measurements of decay constants for Na resonance radiation at Na densities between 10^{13} and 3×10^{16} cm^{-3} have been completed. The density dependences of the decay constant and of the relative intensity of the resonance radiation and of the Na_2 emission are consistent with the collisional transfer of excitation from the $Na(3^2P)$ atoms to the $A^1\Sigma_g^+$ and $a^3\Pi_u$ state of Na_2 and back. This excitation transfer process is helpful in the proposed Na_2 laser scheme since it leads to more rapid formation of excited Na_2 molecules.

II. GENERATION AND INTERPRETATION OF MOLECULAR CONTINUUM RADIATION (Drs. W. P. West and A. Gallagher)

Proposed excimer laser systems utilizing metal vapor-rare gas mixtures are expected and operated at rare-gas pressures of about 10 atm. At lower pressures the gain and absorption coefficients of the medium are primarily determined by the diatomic excimers (AX) and dimers (A_2) where X is the noble gas and A may be any atomic specie. However, the triatomic excimer AX_2 can also make a significant contribution to the gain and absorption coefficients at these high X pressures. During this report period absolute measurements of fluourescent intensity and predictions of stimulated emission coefficients have been made for Na in Xe at the high pressures characteristic of proposed visible lasers. Previously estimated values were obtained by extrapolating measurements made at pressures of one atmosphere and less. The absolute data was obtained by extending the fluorescence measurements to include the spectral region near line center as shown in Fig. 1. These measurements were carried out using a tunable dye laser to excite one of the sodium resonance doublets while measuring the spectral distribution of fluorescence from the other.

The line center measurements made possible integration of the relative fluorescent intensity over the complete spectral range and resulted in the normalization of the far wing spectrum as shown in Fig. 2. As discussed in the last report the fluorescent intensity and predicted stimulated emission cross section for λ > 700 nm is dominated by $NaXe_2$ emission.

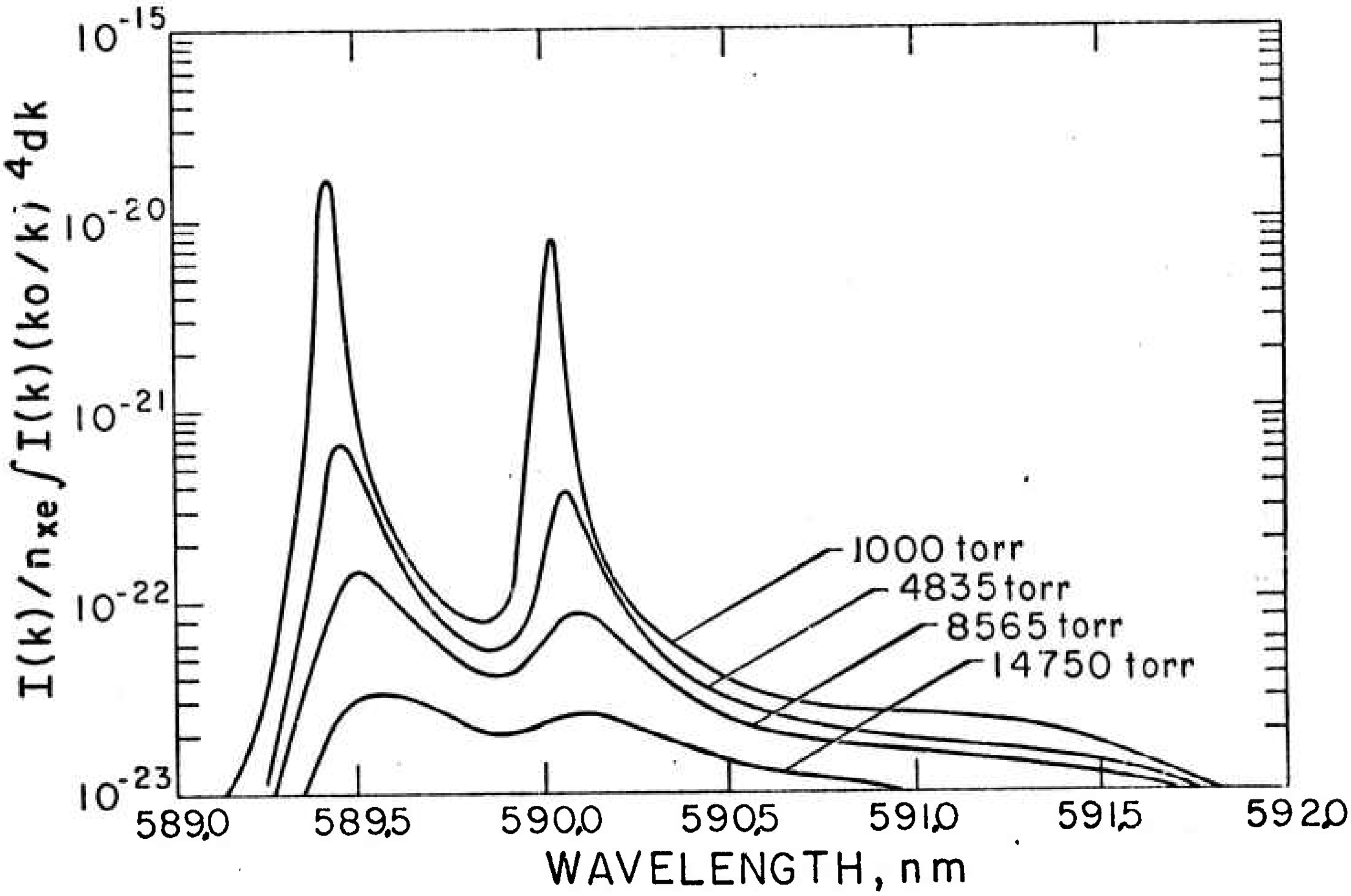

Fig. 1. Normalized fluorescent intensity near line centers for 589.0 and 589.6 nm. The spectrometer wavelength scale is displaced by about 0.4 nm.

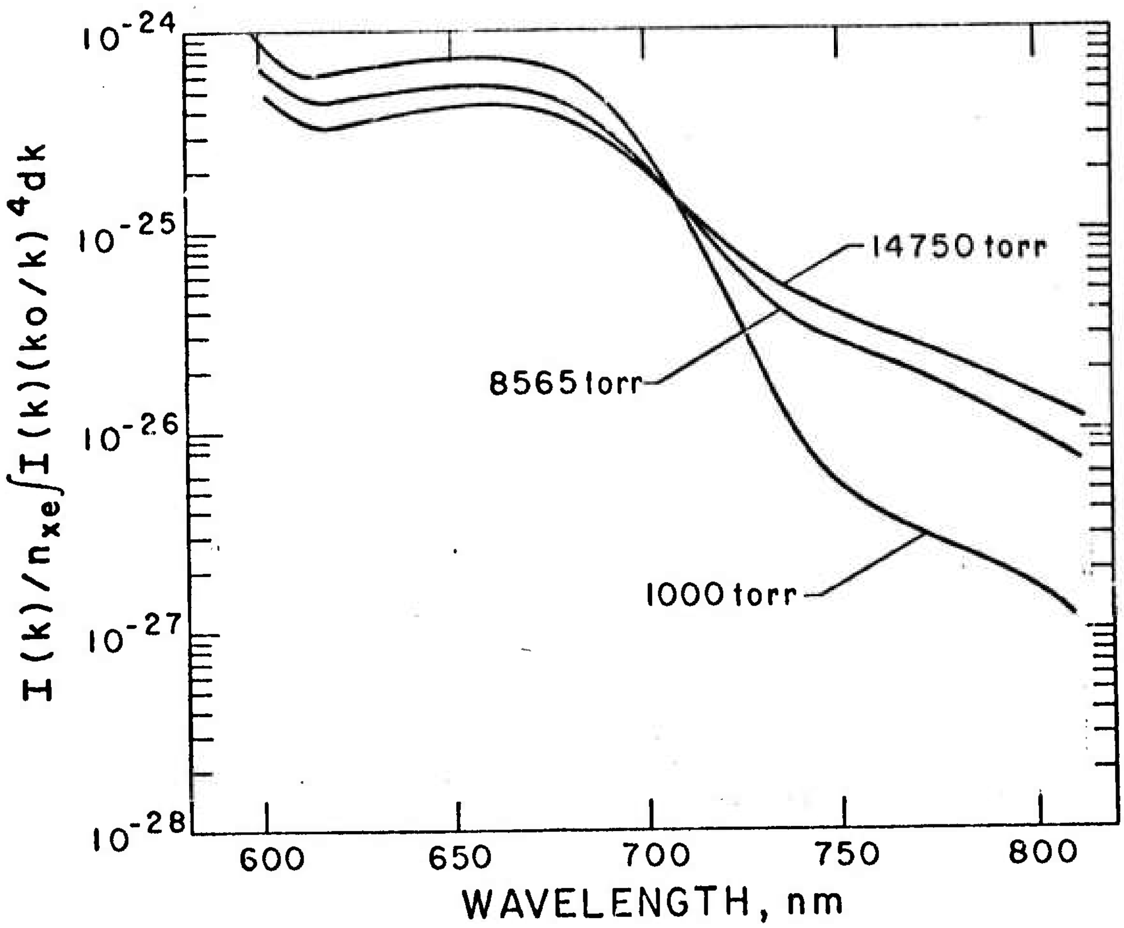

Fig. 2. Normalized fluorescent intensity in red wings of Na resonance lines.

III. STABILITY OF DISCHARGES IN WEAKLY IONIZED GASES (Drs. H.-C. Chen and A. V. Phelps).

The objective of this theoretical investigation of the growth of instabilities in weakly ionized gas discharge is to develop quantitative techniques for the prediction of conditions for formation of the arcs which limit the power output from high power laser systems. The availability of such techniques would greatly aid the optimization of high power lasers cureently under development and would greatly improve the reliability of the design of scaled-up versions of these lasers.

Recent work has been concerned with the development of models and calculation techniques for describing the cathode fall region of electrical discharges such as those being developed for high power, visible lasers. This emphasis was dictated by the large amount of compute storage and time required for this region when attempts were made to model the complete discharge.

Typical results of calculations for a one-dimensional cathode fall model are shown in Fig. 3. Curves of electron and ion density, electric field and electric potential are shown for an electrode spacing of 3.4 cm and a helium pressure of 1 atmosphere. Processes included in the model are electron impact ionization, electron-ion recombination, electron diffusion and drift, ion drift, and secondary electron emission caused by the arrival of ions and photons at the cathode. Figure 3 shows the buildup of electric field at the cathode and the peaks of ion and electron density which advance to the cathode to produce the space charge sheath and region of ionization called the cathode fall.

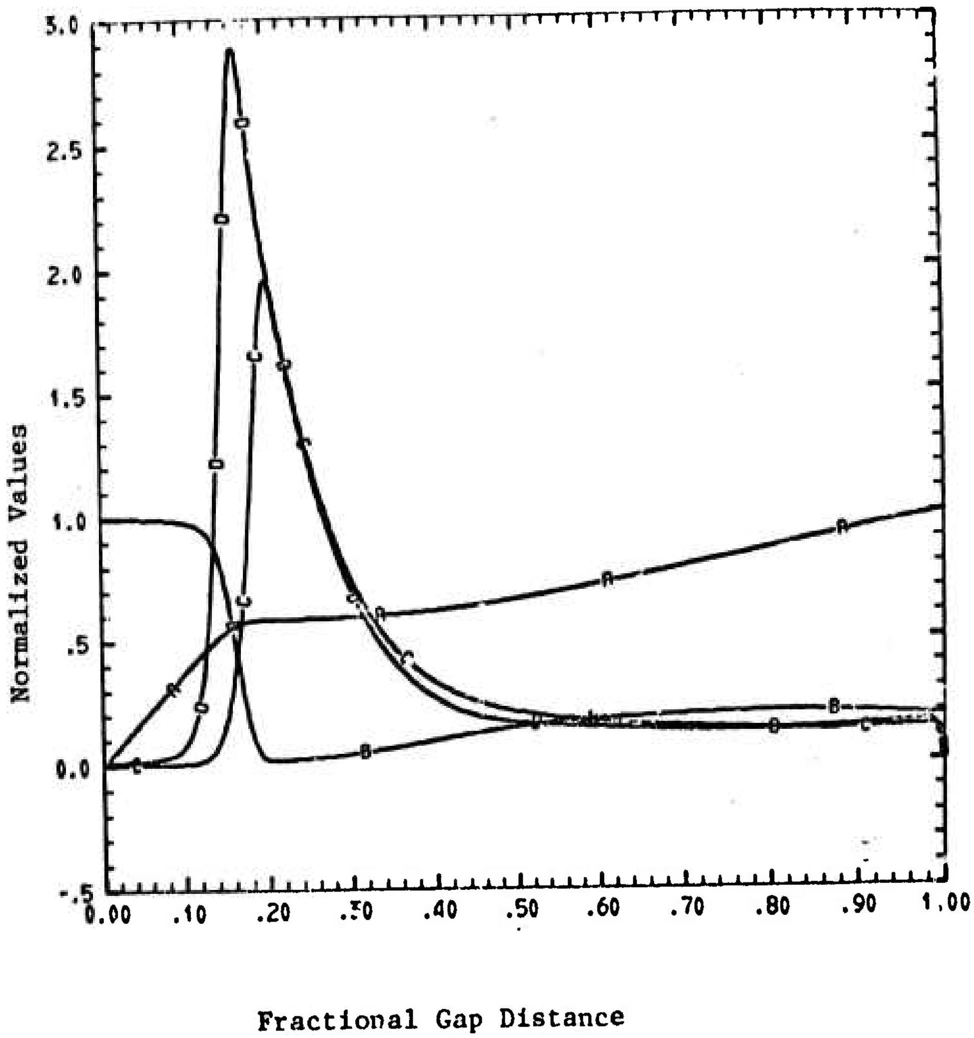

Fig. 3. Calculated field and density distributions for cathode region of a discharge in helium. $E/N|_o = 1.8 \times 10^{-16}$ V-cm^2, d = 3.4 cm, N = 3×10^{19} cm^{-3}, t = 0.14 μsec. The
The curves are: A - electric potential relative to gap voltage;
B - electric field strength in units of 20 kV/cm;
C - electron density in units of 10^{12} electrons/cm^3;
and D - positive ion density in units of 10^{12} ions/cm^3.

IV. ELECTRON EXCITATION OF MOLECULAR METASTABLES (Drs. S. A. Lawton and A. V. Phelps).

The objective of this project is the measurement of excitation and destruction rate coefficients for metastable states of molecules which serve as upper states of lasers or which make possible efficient excitation of such laser states. The measurements of rate coefficients for the electron excitation of O_2 molecules to the $b^1\Sigma_g^+$ state have been completed and are shown in Fig. 4. The range of E/N covered is from 5×10^{-17} to 2×10^{-15} V-cm^2 corresponding to mean electron energies from 0.75 to about 6 eV. At selected E/N values and O_2 densities measurements have been made of the fraction of the charge crossing the drift tube as electrons so as to allow correction of the data for attachment and ionization. At these E/N values, the effective excitation coefficient was determined as a function of O_2 density so as to allow selection of data for which the photoelectrons have reached equilibrium with the applied electric field.

The solid curve of Fig. 4 shows the rate coefficients predicted using the results of recent electron beam measurements[1] and solutions of the Boltzmann equation using our recommended[2] set of electron collision cross sections for O_2. The measured electron excitation rate coefficients are in agreement with predictions for mean energies below 1.5 eV but at the highest mean energies they are about a factor of 30 higher than expected for direct excitation of the $b^1\Sigma_g^+$ state. Present indications are that all of the excited molecules and dissociation products with sufficient energy to produce $b^1\Sigma_g^+$ molecules are collisionally deexcited to the $b^1\Sigma_g^+$ state in a time short compared to the $b^1\Sigma_g^+$ decay time. Thus, the dashed curve of Fig. 4 shows the sum of the excitation rate coefficients for the $b^1\Sigma_g^+$

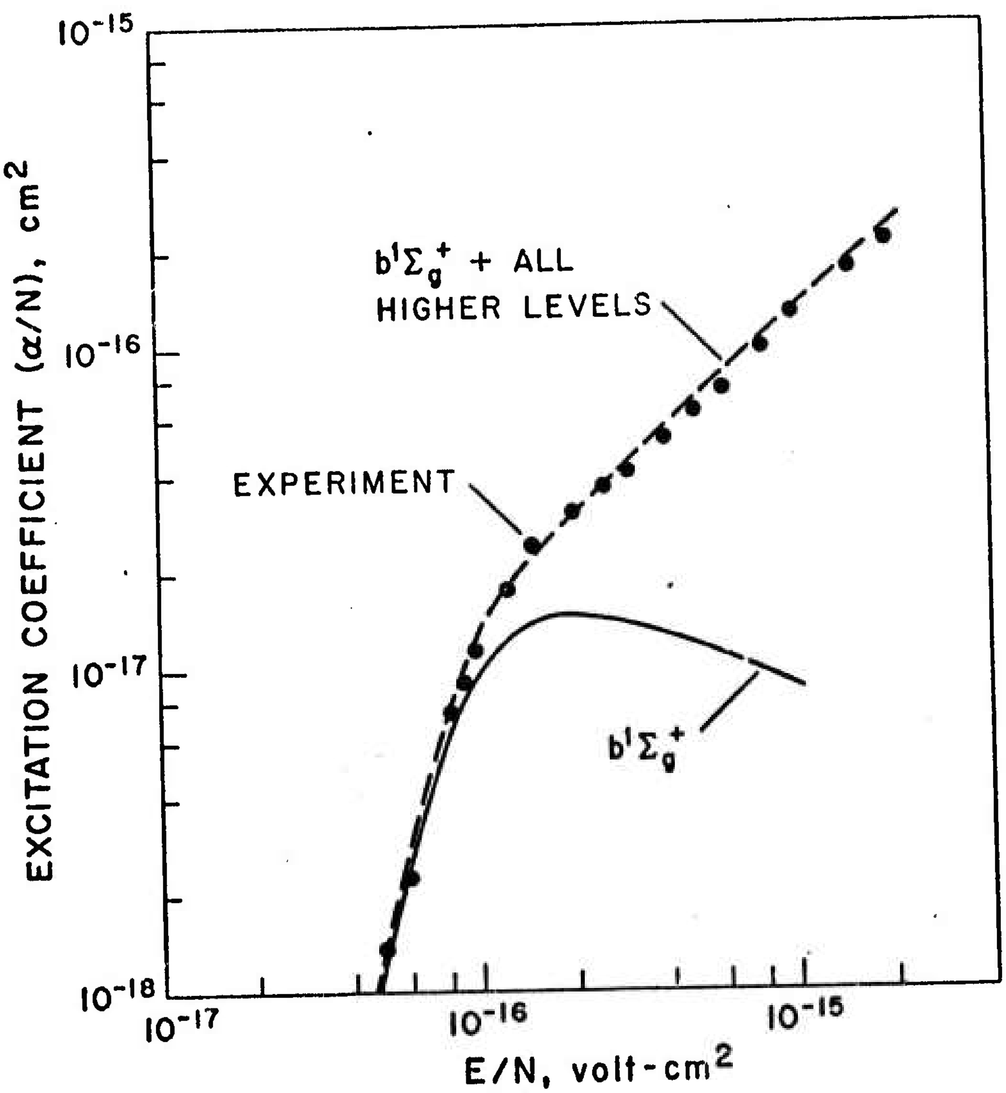

Fig. 4. Measured excitation coefficients for 762 nm band of O_2. Solid curve is prediction for direct excitation of $b^1\Sigma_g^+$ state. Dashed curve is sum of excitation rate coefficients for $b^1\Sigma_g^+$ and all higher electronic states of O_2 as calculated using our recommended cross section set.

state, the 6 eV states ($c^1\Sigma_u^-$, $C^3\Delta_u$ and $A^3\Sigma_u^+$), and for the $B^3\Sigma_u$ state. In the case of the $B^3\Sigma_u^-$ or Schuman-Runge state, the process of $O(^1D)$ formation and sbusequent excitation transfer to the $b^1\Sigma$ state is well known.[3]

V. SCATTERING AND TRANSPORT OF RESONANCE RADIATION (Drs. T. Fujimoto and A. V. Phelps).

The objective of this project is to determine the rates of radiative and non-radiative energy loss from resonance states of a typical metal vapor atom (Na). Such data is essential to the accurate modeling of proposed high power metal vapor-rare gas excimer lasers operating at near visible wavelengths In colloaboration with Drs. L. Lam and M. M. Hessel at NBS/Boulder, we have imitiated a series measurements of the decay of resonance radiation in Na vapor at high Na densities so as to yield more directly the rate coefficients for collisional energy transfer and quenching in Na vapor. A pulsed dye laser tuned to wavelengths in the vicinity of the 590 nm resonance lines of Na was used to excite the Na (3^2P) states. Measurements were then made of the time variation of the 590 nm atomic radiation and of the molecular intensity in the 590 to 800 nm range. The Na density was varied from 10^{13} to 3×10^{16} cm^{-1}. At the higher Na densities the vapor temperature was varied from 440 to 480 C.

Figure 1 shows the variation of the decay constants for the atomic and molecular radiation and of the ratio of the total intensity of molecular radiation to the total intensity of atomic radiation. The points are the experimental data and the smooth curves are theoretical curves calculated using a model including the following processes:

1) The transport of resonance radiation. Here the theory of Holstein[4] for self-broadened lines predicts a loss rate which is independent of Na density so long as the resonance line has a Lorentzian shape.

2) Excitation transfer by the process $Na(3^2P) + Na_2(X^1\Sigma_g) \rightarrow Na_2(A^1\Sigma_g^+$ and $a^3\Pi_u) + Na(3^2S)$. The large rate coefficient derived

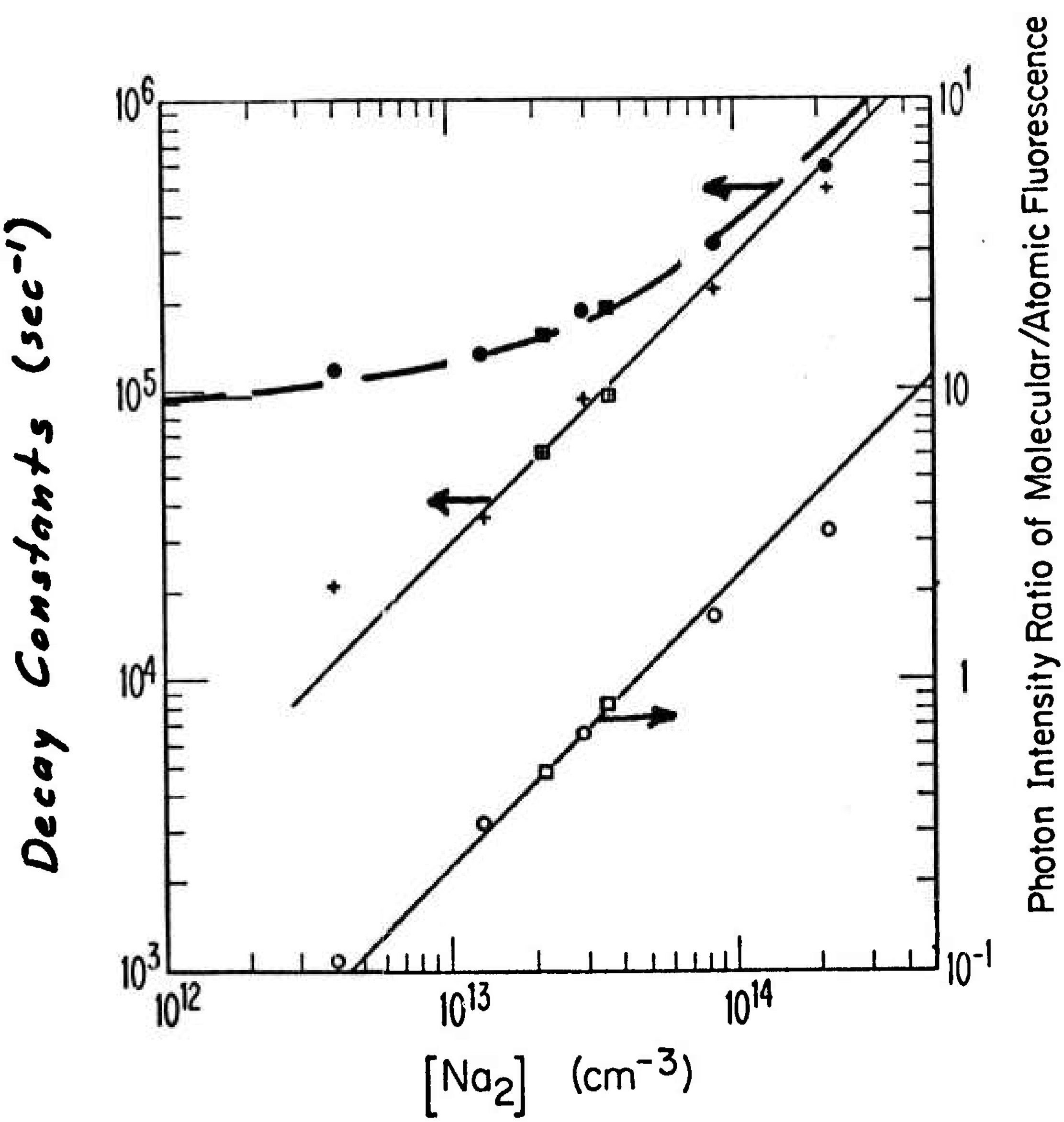

Fig. 4. Decay constants and molecular to atomic intensity ratios vs Na_2 density. The solid circles and squares are measured final decay instants at 420 ± 10 C and 440 C, respectively. The crosses and boxed crosses are the measured decay constants minus the theoretically predicted decay constant for the collision broadened resonance radiation. The open circles and crosses are the measured intensity ratio. The smooth curves are the predictions of our model when only processes 1) and 2) of the text are included. The departures of experiments from these curves at the higher Na_2 densities are indicative of processes 3) through 5).

for this process (3×10^{-9} cm^3/sec) suggests the importance of a long-range interaction between excited Na and Na_2. In pure sodium and in Na-rare gas discharges at low total pressures, e.g., 1 atmosphere, this process is expected to provide the major route for the formation of excited Na_2 from the excited Na produced by electron excitation.

3) Excitation transfer from excited Na_2 to $Na(3^2P)$ atoms, i.e., the reverse of process 2). This process has recently been observed at low Na densities by Kraulinya _et al_. Our estimates of the rate coefficient for this process are dependent upon estimates of an effective radiative lifetime for the $a^3\Pi_u$ state.

4) Radiative transitions from vibrational-rotational levels of the $a^3\Pi_u$ state of Na_2 to the ground state $X^1\Sigma_g^+$. The estimates of the transition probabilities for this are based on studies of the perturbations of levels of the $A^1\Sigma_u^+$ state by levels of the $a^3\Pi_u$ state by Hessel and Kusch.[6]

5) Excitation transfer among the $A^1\Sigma_u^+$ and $a^3\Pi_u$ states, vibrational relaxation within these states, and quenching of the $a^3\Pi_u$ state by predissociation and collisions. Attempts are being made to improve the estimates of these rate coefficients.

REFERENCES

1. S. Trajmar, D. C. Cartwright and W. Williams, Phys. Rev. A4, 1482 (1971); F. Linder and H. Schmidt, Zeit. für Naturforsch 26a, 1617 (1971); R. I. Hall and S. Trajmar, J. Phys. B 8, L293 (1975).
2. A. V. Phelps and S. A. Lawton, submitted to 1976 Gaseous Electronics Conference.
3. D. W. McCullough and W. D. McGrath, Chem. Phys. Letters 8, 353 (1971).
4. T. Holstein, Phys. Rev. 72, 1212 (1947); 83, 1159 (1951).
5. E. K. Kraulinya, E. K. Kopeikina and M. L. Janson, Chem. Phys. Letters 39, 565 (1976).
6. M. M. Hessel and P. Kusch (unpublished).